Micro/atomic-scale vibration induced superlubricity

Shuai SHI^{1,2}, Dan GUO^{1,*}, Jianbin LUO^{1,*}

¹ State Key Laboratory of Tribology, Tsinghua University, Beijing 100084, China

² China Fortune Land Development Industrial Investment Co., Ltd., Beijing 100027, China

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Abstract: With the rapid development of industry, the inconsistency between the rapid increase in energy consumption and the shortage of resources is becoming significant. Friction is one of the main causes of energy consumption; thus, the emergence of superlubricity technology can substantially improve the energy efficiency in motion systems. In this study, an efficient method to control superlubricity at the atomic-scale is proposed. The method employs vibrational excitation, which is called vibration induced superlubricity (VIS). The VIS can be easily and steadily achieved by employing external vibration in three directions. The simple method does not depend on the type of sample and conductivity. Dependence of oscillation amplitude, frequency, scanning speed, and normal force (F_N) on friction were investigated. Experimental and simulated explorations verified the practical approach for reducing energy dissipation and achieving superlubricity at the atomic-scale.

Keywords: superlubricity; atomic-scale; vibration; energy dissipation

1 Introduction

Energy losses due to friction and wear are estimated to account for 23% of the world's total energy consumption [1]. The reduction of friction is not only a problem to be solved in engineering applications, but also a technical barrier for micro-devices such as micro-electromechanical systems. New technologies for lubrication and materials related to the energy-saving and green industry are urgently required. Currently, some advanced solutions have been proposed to potentially solve friction and wear problems for micro- or nano-electro-mechanical systems (MEMS/NEMS) [2–6]. Technologies for the ultra-low friction coefficient (COF) even for superlubricity can greatly improve the energy efficiency of the relative motion system. Superlubricity occurs when COF is lower than 0.01 [7–14]. It was first proposed by Hirano and Shinjo [15] in the early 1990s. It has been studied by various groups, and the relevant mechanism of

energy consumption has also been explored [16–18].

There are two main mechanisms responsible for superlubricity. One is due to the nature of the material itself and the other is due to external conditions. Superlubricity may occur when two crystalline surfaces slide over each other in dry incommensurate contact [19–22]. This effect, which is called structural lubricity, was suggested in 1991 and verified with great accuracy using two graphite surfaces. Dienwiebel et al. [21] observed superlubricity while turning a graphite flake out of registry over a graphite surface in 2004. Socoliuc et al. [23] experimentally demonstrated and theoretically described a state of ultra-low dissipation in atomic friction. The friction can be controlled by varying the load on a nanometer scale contact. In 2006, Socoliuc et al. [24] proposed a new method to achieve superlubricity by adding voltage to the conducting probe when scanning ionic crystal by atomic force microscopy (AFM). They found that friction energy consumption

* Corresponding authors: Dan GUO, E-mail: guodan26@tsinghua.edu.cn; Jianbin LUO, E-mail: luojb@tsinghua.edu.cn

almost disappeared when the voltage frequency was applied. Riedo et al. [25] applied lateral oscillations to the cantilever holder while sliding on mica. They found a reduction in the average friction force around the resonance frequency when the sliding velocity was below the critical value. Adjusting sliding speed and thermal excitation can also be used to reduce friction force (F) [26].

Unfortunately, superlubricity cannot be easily achieved in these techniques and applied in practical engineering applications. Some methods need accurate control of relative orientation while some others need complex conditions for specific materials. However, if the realization of superlubricity depends on the conductivity of the sample, the possibility of having a universal method would not be high enough. In the present study, superlubricity was achieved by employing an atomic-scale innovative method using AFM. Applying additional vibration to the probe under constant F_N during the frictional process is the concept of vibration induced superlubricity (VIS). In this study, motions of two contacting bodies were modulated at well-defined frequencies by external vibrations. Superlubricity state was steadily achieved on highly oriented pyrolytic graphite (HOPG) surface. The dependence of amplitude and frequency of vibration on the mean F was explored. Vibration induced superlubricity derived from x - or y -directional vibration on the micro/atomic-scale, which is not commonly explored, was also implemented. The mechanisms of VIS in vertical and lateral directions were investigated.

2 Experimental

2.1 Vibration induced friction force mode (FFM) and calibration

A commercial AFM (Bruker's ICON series, USA) and the FFM were employed in the experiments. A sine voltage signal generated by an additional signal generator was added to the scanning ceramic tube to achieve superposition of movement. The piezoelectric ceramic vibration drove the probe to produce simple harmonic vibration, and thus achieving vibration inducement during the friction process. This method can be used to control friction by adding a vibration in the z -, x -, or y -directions of the

piezoelectric ceramic tube. It is applied to reduce friction even for superlubricity at the atomic-scale. The schematic of vibration induced FFM is shown in Fig. 1.

A commercial probe (AC 240, Olympus, Japan) was used to carry out the friction experiments on the HOPG surface. A thin aluminum film was coated on the Si cantilever to reflect light from the deflection sensor. The tip radius was 7 nm. The F_N and F were monitored through the normal and lateral changes of the laser on the position-sensitive detector (PSD) [27]. Thermal noise power spectra and improved wedge methods were employed to calibrate the cantilever [28–31]. The longitudinal sensitivity coefficient of the probe, the resonant frequency (f_0), the quality factor (Q), and the rigidity coefficient (k) of the probe were 157 nm/V, 61.7 kHz, 132, and 3.174 N/m, respectively. There was a direct relationship between lateral sensitivity coefficient (β) and F_N . Table 1 shows the calibration values for β corresponding to various F_N values. Figure 2 shows the microscopic change of friction process between probe tip and sample after adding additional vibration.

2.2 Micro-scale VIS

Vibration excited friction experiments were conducted on the HOPG surface. Amplitude and frequency were the main parameters first considered for their effects on friction. The other parameters, such as pressure

Fig. 1 Schematic of vibrational excited FFM. A sine voltage signal generated by an additional signal generator was added to the scanning ceramic tube in FFM mode.

Table 1 Calibration values for lateral sensitivity coefficient corresponding to various F_N values.

F_N (V)	1	1.5	2	2.5	3	3.5	4	4.5	5
β (nN/V)	4,827	3,576	3,333	3,346	3,191	3,019	2,905	2,809	2,810

Fig. 2 Additional vibration forces at the tip to vibrate during normal friction movement. The F_N was monitored using a four-quadrant photodetector and controlled by a real-time feedback circuit. v is the scanning speed.

of 498.6 nN, scanning range of 500 nm × 500 nm, and scanning speed of 1 Hz, were also fixed. Firstly, the influence of amplitude and frequency on friction produced on the HOPG surface was investigated. Vibration frequencies in z direction were 1, 2.5, 5, 10, 30, 61.7, and 100 kHz while the amplitude ranged from 0 to 16 nm. Figure 3 shows the curves for friction produced by sliding the probe on the HOPG surface under different vibration amplitudes and frequencies. It can be seen that friction decreased rapidly with the increase in amplitude and tended to flat at 0.5 nN for each vibration frequency. When the vibration frequency was 5 kHz, the friction decreased the fastest.

It is not only the amplitude and frequency of vibration that influence friction, but also the scanning speed. The influence of scanning speed on friction was also studied on the HOPG surface. In the experiments, the scanning speed was discretely selected (0.5, 1, and 2 Hz). The z -directional frequency was 1 kHz while the vibration amplitude ranged from 0 to 16 nm. Figure 4 shows the friction, which resulted from sliding the probe on the HOPG surface at different scanning speeds. It can be seen that as the amplitude increased, the friction at each scanning speed decreased abruptly. It reduced from 0.99 to 0.2–0.3 nN for 0.5 Hz, from 1.75 to 0.3–0.4 nN for 1 Hz, and from 2.61 to 0.7–0.8 nN for 2 Hz. As friction on the graphite surface is relatively small, the initial

Fig. 3 Friction (average frictional force) detected through scanning and z -directional excitation on the HOPG surface as a function of the vibration amplitude (0–16 nm) for different vibration frequencies (1, 2.5, 5, 10, 30, 61.7, and 100 kHz).

friction is mainly related to the position of the tip and the degree of wear. Thus, fluctuation in initial friction is normal. When the scanning speed was large, the friction without vibration was also large. This indicates that scanning speed has a substantial influence on friction stability. However, it was evident that friction decreased significantly with additive vibration. The faster the scan, the greater the friction.

It is well known that macroscopic friction is proportional to normal load. However, for micro/atomic-scale, the actual contact area between two surfaces is several orders of magnitude smaller than the apparent contact area. The change in F_N also affects the contact state. In the study, the relationship between F_N and friction was investigated experimentally on a HOPG surface. A scanning speed of 1 Hz, a vibration frequency of 1 kHz, and vibration amplitudes of 0, 0.4, 1, 1.4, 2, 2.4, and 3 nm were employed in the experiments. Figure 5 shows the experimental results for the change of friction with the F_N for the different vibration amplitudes. The COF was obtained by fitting the discrete point of each vibration amplitude. An amplitude of 0 indicates that there is no vibration inducement. In this case, it is caused by the normal friction mode.

Fig. 4 Friction detected by scanning and z -directional vibration excitation on the HOPG surface as a function of the amplitudes (0–16 nm) for the different scanning speeds (0.5, 1, and 2 Hz).

Fig. 5 Friction on the HOPG surface as a function of the applied normal load for different z -directional amplitudes (0, 0.4, 1, 2.4, and 3 nm) with a vibration frequency of 1 kHz.

The COF (μ_0) was 0.0123 when the amplitude is 0. As the F_N increased, F also increased correspondingly. When the vibration amplitude was 0.4 and 1 nm, F increased with the increase of F_N . However, it was significantly slower than that without the addition of vibration. After fitting, $\mu_{0.4} = 0.0078$, $\mu_{1.0} = 0.0017$, $\mu_{1.4} = 0.0004$, $\mu_2 = 0.0012$, $\mu_{2.4} = 0.0016$, and $\mu_3 = 0.0007$ were obtained for the corresponding vibration amplitudes of 0.4, 1.0, 1.4, 2, 2.4, and 3 nm, respectively. In comparison, friction can be significantly reduced and the COF can be greatly flattened with additive vibration. The COF reached one-thousandth of a magnitude, indicating superlubricity was achieved.

As the friction with z -directional vibration has been studied, it was also important to determine whether the friction with x - or y -directional vibrations would also decrease. Similarly, the selected sine voltage signal generated by an extra signal generator was added to the x or y direction of the piezoelectric ceramic tube. It can perform common friction test for x - or y -directional vibration. Friction behaviors with x - or y -directional vibration on the HOPG surface were also investigated. Probe AC240, 1 Hz scanning speed, 5 kHz vibration frequency, and 3 V (1,334 nN) F_N were employed with x -directional vibration amplitude of 0–10 nm (0–11 nm in the y direction). Figure 6 shows the change in friction with vibration amplitude in x or y direction. It can be seen that with an increase in amplitude, friction decreased rapidly with x - or y -directional vibration excitation. Friction decreased and gradually flattened at approximately 0.5 nN for both x - and y -directional vibrations. Therefore, additive vibration excitation is a common method for reducing friction in graphite surfaces in the three different directions. Compared with other related studies, friction associated with x - or y -directional vibration has never been previously studied in detail; thus, the significant reduction in friction indicates its importance in achieving superlubricity.

2.3 Atomic-scale VIS

To explore the mechanism of the VIS, atomic-scale friction experiments were further carried out on graphite using an Olympus probe AC240. The coefficients of the transverse sensitivity are shown in Table 2. The longitudinal sensitivity coefficient of the probe was 133.8 nm/V, f_0 was 78.8 kHz, Q was 183, and k of the probe was 3.45 N/m. Table 2 shows the calibration values for β corresponding to various F_N values.

Initially, atomic-level friction was produced on the HOPG surface through the normal friction mode. An image of atomic-level friction is shown in Fig. 7. The real-time trace and retrace curves corresponding to the atomic-level stick-slip curves can be seen in Fig. 7(a). The middle region of the trace and retrace curves represents twice the voltage value corresponding to the F . Friction ring (friction

Fig. 6 Friction detected by scanning on the HOPG surface as a function of vibrational amplitudes for (a) *x*- and (b) *y*-directional vibration.

Table 2 Calibration values for lateral sensitivity coefficient corresponding to various F_N values.

F_N (V)	0.5	0.75	1	1.25	1.5	1.75	2	2.25
	2.5	2.75	3	3.25	3.5	3.75	4	—
β (nN/V)	1,996	2,260	2,365	2,550	2,605	2,658	2,622	2,611
	2,680	2,676	2,640	2,605	2,593	2,576	2,544	—

Fig. 7 Friction images detected by scanning forward and backward on an atomically flat HOPG surface: (a) Without *z*-directional vibration and (b) a comparison between *z*-directional vibration and no vibration.

energy consumption) disappeared with the addition of *z*-directional vibration and appeared when disconnected to a vibrational signal as shown in Fig. 7(b). It can be clearly seen from the comparison that VIS can be easily achieved.

Amplitude and frequency are still the main

parameters that may influence the results for friction at the atomic-scale. In the experiment, the value for F_N was 1 V (460.6 nN), the scanning range was 5 nm × 5 nm, the scanning speed was 1 Hz, the frequencies of *z*-directional vibration were 1 and 5 kHz, and the amplitude was 0–4 nm. Figure 8(a) shows the changes

in friction caused by the slipping probe on the atomic-scale HOPG surface under different vibration amplitudes and frequencies. Friction decreased rapidly with the increase in amplitude and tended to be close to 0 both for 1 and 5 kHz. This indicates that the superlubricity state was achieved on the atomic-scale HOPG surface with additive vibration. Similarly, experiments were conducted to investigate the influence of frequency on atomic-level friction. Vibration

amplitude was set to be 0.2 and 0.6 nm. As shown in Fig. 8(b), friction drops rapidly and stays close to 0 when the vibration signal was used to modulate the scanner. However, friction did not reduce significantly under 10–15 kHz and the amplitude affected the level of reduction. Additionally, there was no apparent decrease in friction under 30–40 kHz. Nevertheless, the range of frequencies that can achieve superlubricity is relatively wide.

Fig. 8 Friction detected by scanning on the atomic-scale HOPG surface as a function of the (a) z -directional vibration amplitude at 1 and 5 kHz frequencies, (b) z -directional vibration frequency at 0.2 and 0.6 nm amplitudes, (c) x -directional vibration amplitude at 1 and 5 kHz frequencies, (d) x -directional vibration frequency at 0.6 nm amplitude, (e) y -directional vibration amplitude at 1 and 5 kHz frequencies, and (f) y -directional vibration frequency at 0.5 nm amplitudes.

Similarly, the friction experiments with x - and y -directional vibration on the graphite were conducted. For x -directional vibration excitation, the value for F_N was 1 V (460.6 nN), the scanning range was 5 nm $\times$ 5 nm, the scanning speed was 1 Hz, the vibrational frequencies were 1 and 5 kHz, and the vibration amplitude ranged from 0 to 10 nm. Figure 8(c) shows the variation of atomic-level friction with the x -directional vibration. Friction decreased faster and tended to be flat near 0 when the amplitude was increased under a 1 kHz frequency. Friction also shows dependence on the frequency band in the x direction at an amplitude of 0.6 nm as shown in Fig. 8(d). Friction can be generally reduced at a frequency band of 0–10 kHz and even at some points within 10–40 kHz.

In the y direction, atomic-level experiments for friction on the HOPG surface were also conducted at 0–4.5 nm vibrational amplitude. The changes in atomic-level friction are shown in Fig. 8(e). When the vibrational frequencies were 1 and 5 kHz, friction decreased rapidly with an increase in amplitude and tended to be 0. Figure 8(f) shows that friction decreased at 0–10 kHz and 17–33 kHz, and 0.5 nm vibration amplitude. However, no substantial decrease was observed at 10–17 kHz.

The effect of scanning speed on atomic-level friction was also investigated. Figure 9 shows the resulting friction caused by AC240 probe sliding on the atomic surface of HOPG at scanning speeds of 0.5, 1, and 2 Hz. Figure 9(a) shows the friction in the z direction at a frequency of 1 kHz and discrete change of vibrational amplitude of 0–4 nm. Friction without additive vibration under scanning speeds of 0.5, 1, and 2 Hz were 23.9, 25.1, and 26.7 nN, respectively. As the amplitude increased, friction abruptly decreased and gradually flattened between 0 and 0.04 nN for each speed. Similarly, in Fig. 9(b), values for friction without vibration for speed of 0.5, 1, and 2 Hz were 15.1, 17.8, and 19 nN, respectively, and abruptly decreased to approximately 0.01 nN with x -directional vibration (vibrational frequency of 1 kHz and amplitude of 0–5 nm). Values for friction without vibration at 0.5, 1, and 2 Hz were 9.26, 12, and 13.7 nN, respectively, and decreased suddenly to approximately 0.03 nN with y -directional vibration (vibrational frequency of 1 kHz and amplitude

of 0–4.5 nm) as shown in Fig. 9(c). It can be seen that friction at small scanning speeds decreased slightly and the atomic-level friction in the x , y , and z directions could achieve the VIS state.

Fig. 9 Friction detected by scanning on the atomic-scale HOPG surface as a function of amplitude (0–5 nm) for the different directional vibrations: (a) z -directional, (b) x -directional, and (c) y -directional vibrations.

As shown in Fig. 10, changes in friction by sliding the probe on the atomic HOPG surface at different normal forces were obtained. When additive vibration (0 nm amplitude) was not applied, friction increased with increasing F_N ; the COF was approximately 0.006. Figure 10(a) shows the curves for friction at

Fig. 10 Friction as a function of the applied normal load for different directional vibrations: (a) z -directional, (b) x -directional, and (c) y -directional vibrations.

z -directional vibration frequency of 1 kHz and amplitudes of 0.2, 0.5, and 1 nm. When the additive vibration was applied, there was nearly no change in friction as F_N was increased. As can be seen in the enlarged detail diagram from Figs. 10(a)–10(c), the fluctuation distribution of COF was less than 0.006. The COFs for x - and y -directional vibrations at a frequency of 1 kHz and amplitudes of 0.2, 0.6, and 1 nm are shown in Figs. 10(b) and 10(c), respectively. The average COF was less than 0.006. This indicates the vibration-induced friction state of superlubricity.

3 Numerical results and discussion

Analytical or numeric calculations can be employed to simulate the behavior of atomic friction. Prandtl–Tomlinson (PT) model is a simple but widely used model, which consists of a harmonic spring and an equivalent point-mass. The models were established to simulate the influence of vibration inducement on friction. Mechanisms of vibration-induced superlubricity in z and x directions were investigated. The schematic of a one-dimensional (1D) PT model is shown in Fig. 11.

The cantilever-tip system is generally approximated as a single degree of freedom damped spring oscillator model. The total potential energy of the system $V(x, t)$ can be written as

$$V(x, t) = -\frac{U}{2} \cos\left(\frac{2\pi x}{a}\right) + \frac{1}{2}k(vt - x)^2 \quad (1)$$

Fig. 11 Schematic of a 1D PT model, showing the relationship between AFM tip/cantilever and sample. Rectangular block (sliding support) leads the red solid sphere (tip atoms) through a corrugated potential energy landscape. f_v and f_l are vertical and lateral oscillation.

$[U\cos(2\pi x/a)]/2$ describes the corrugation potential where U is the magnitude of the corrugation potential and a is the lattice spacing of the sample. $k(vt-x)^2/2$ is the elastic potential interaction between the tip and support linked by a spring whose constant is defined as k . x is the coordinate of the tip atom. Besides, vt is the position of the support, t is the scanning time. The dynamics of the tip can be described by the Langevin equation [32]:

$$m\ddot{x} + m\mu\dot{x} = -\frac{\partial V(x,t)}{\partial x} + f_r \quad (2)$$

where m is the mass of the tip, μ is the viscous friction (or damping) coefficient, and f_r is a random force field which satisfies the fluctuation–dissipation relation in the stochastic process.

$$\langle f_r(t)f_r(t') \rangle = 2m\mu k_B T \delta(t-t') \quad (3)$$

$\langle \rangle$ indicates an ensemble average, k_B is the Boltzmann constant, T is the absolute temperature, and δ is the Dirac delta function. The F on a tip atom was calculated using the 1D Tomlinson model at a finite temperature. The fourth-order Runge–Kutta (RK) algorithm developed by Kasdin was used to numerically solve the Langevin equation [33]. The F is given by

$$F = k(vt - x) \quad (4)$$

3.1 Normal VIS

The variation of vertical position can be implicitly reflected by the variation of U [34]. The magnitude of the corrugation potential has a harmonic shape when U is supposed to be linear with the vertical position.

$$U = U_0 [1 + \alpha \sin(2\pi f_v t)] \quad (5)$$

where U_0 is the initial amplitude of corrugation potential, α and f_v are the amplitude and frequency of superimposed vertical oscillation, respectively.

The dependence of time-mean F on α and f_v (unit: $\sqrt{k/m}$) are demonstrated in Fig. 12(a). Greater vibrational amplitude and lower excitation frequency could decrease the atomic friction significantly. For high frequencies, the tip atom could not follow the vertical oscillation of the support motion. In

Fig. 12 Dependence of mean F_N on α and f_v : (a) 2D map and (b) dependence details of friction on f_v and α .

addition, the fluctuation of the tip atom could not jump from one potential well to the next when the vibrational amplitude was small. Thus, the mean F mostly remained constant as there was no vibration. Transverse lines of Fig. 12(a) depict the dependence details of friction on f_v and α in Fig. 12(b). There were two local friction minima in every $F-f_v$ curve. The first minimum occurred from the resonance between lattice frequency v_0/a and external excitation frequency f_v . They were nearly located at the same position for all $F-f_v$ curves. The second minimum occurred when it produced resonance between f_v and frequency of PT mode. The slip position influenced by the frequency of PT mode may determine the location of the second minimum.

Comparing the experimental results in Fig. 3, friction showed a significant reduction when the frequency was 5 kHz. Even if the amplitude was relatively small, it still showed an ideal superlubricity effect. Based on

the analysis of the simulation, it occurred from the resonance between lattice frequency and external excitation frequency f_v . At this resonance point, the contact area was supposed to be the minimum. In addition, it can be seen that friction also showed a significant reduction when the frequency was 61.7 kHz. The effect was more likely to occur because of the resonance between the external excitation frequency f_v and the scanning system. For 1 and 100 kHz, the reduction in friction was not significant and fast. The experimental results and simulation trends were very consistent. Appropriate selection of frequency is crucial for achieving a significant reduction in friction and superlubricity.

3.2 Lateral VIS

Excitation of the lateral mode leads to an oscillating motion of the tip in the scanning direction. The oscillating motion can be implicitly reflected by variation of support position [35]. In this case, the position of the support in the PT model as shown in the following expression:

$$vt = v_0 t + \gamma a \sin(2\pi f_1 t) \quad (6)$$

where γa and f_1 are the amplitude and frequency of superimposed lateral oscillation, respectively.

The dependence of mean F on γa and f_1 (unit: $\sqrt{k/m}$) is shown in Fig. 13(a). Similar to the discussion in vertical vibration situation, greater vibrational amplitude and lower excitation frequency could achieve vibration-induced superlubricity. The tip atom could not follow the lateral oscillation of the support motion at high frequency. For small vibrational amplitude, the motion of the support was not able to lead the tip atom to make meaningful oscillation. Thus, the mean F did not change in these two situations. Figure 13(b) shows the dependence of the mean F on f_1 and γ . There were also two local friction minima in every $F-f_1$ curve. The first minimum occurred from the resonance between lattice frequency v_0/a and f_1 , similar to that for vertical vibration. Similarly, the second minimum occurred when it produced resonance between external excitation frequency f_1 and frequency of PT mode.

Fig. 13 Dependence of the mean F on γ and f_1 : (a) 2D map and (b) dependence details of friction on f_1 and γ .

4 Conclusions

Superlubricity at the atomic-scale can be achieved by introducing an external vibration signal. Friction experiments with x -, y -, and z -directional additive vibration on a graphite surface indicated that the superlubricity state can be steadily implemented through the different directional vibration excitations. The influences of vibration amplitude, frequency, scanning speed, and F_N on mean F were investigated. An appropriate combination of frequency and amplitude is crucial for achieving a significant reduction in friction and superlubricity. This approach can be extended to MEMS/NEMS at the micro/atomic-scale and mechanical wear reduction at the macro-scale.

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Shuai SHI. He obtained his B.E. degree from Northeastern University in 2011, and received his M.E. degree from Huazhong University of Science and Technology in 2014. In 2018, he received his Ph.D. degree from Tsinghua University, China. He is

Dan GUO. She received the M.S. degree in engineering mechanics in 1995 from Xi'an Jiaotong University, Xi'an, China and Ph.D. degree in engineering mechanics in 1999 from Tsinghua University, Beijing, China. She joined the State Key Laboratory

Jianbin LUO. He received his B.Eng. degree from Northeastern University in 1982, and got his M.Eng. degree from Xi'an University of Architecture and Technology in 1988. In 1994, he received his Ph.D. degree from Tsinghua University and then joined the faculty of Tsinghua University. Prof. Jianbin LUO is an academican of the Chinese Academy of Sciences and a Yangtze River Scholar

working at State Key Laboratory of Tribology, Tsinghua University, as well at China Fortune Land Development Industrial Investment Co., Ltd. for postdoctoral research. His research interests include vibration induced superlubricity and energy dissipation at the micro/nano scale.

of Tribology at Tsinghua University from 1999. Her current position is a professor and the deputy director of the laboratory. Her research areas cover the properties of friction at the micro/nano-scale, mechanism of interaction among nanoparticles and surface in ultra-smooth surface planarization, and the formation and failure of lubricant film in harsh conditions.

Distinguished Professor of Tsinghua University, Beijing, China. He was awarded the STLE International Award (2013), the Chinese National Technology Progress Prize (2008), the Chinese National Natural Science Prize (2018, 2001), and the Chinese National Invention Prize (1996). Prof. LUO has been engaged in the research of thin film lubrication and tribology in nanomanufacturing. He has been invited as a keynote or plenary speaker for 20 times on the international conferences.